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On the mechanism of $BaSi_2$ thin film formation on Si substrate by vacuum evaporation

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Abstract

We report on the formation mechanism of $BaSi_2$ thin film on Si substrate grown by vacuum evaporation using $BaSi_2$ granules as source materials. Since the vapor flux at the initial stage of evaporation is known to be Ba-rich, Si supply from the substrate is of crucial importance to obtain homogeneous $BaSi_2$ thin film. In fact, low substrate temperature and/or thick film deposition led to formation of rough film with voids, and the oxidation proceeded upon exposure to air. We revealed that appropriate choice of substrate temperature, film thickness, and post-growth *in-situ* annealing can provide enough diffusion of Si and Ba, leading to realization of homogeneous $BaSi_2$ thin film.

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Keywords: barium desiliside, vacuum evaporation;

1. Introduction

Orthorhombic $BaSi_2$ is attractive material as an absorption layer for single junction thin-film solar cells because it has suitable band gap (1.3 eV [1,2]) and large absorption coefficients (3×10^{-4} cm^{-1} at 1.5 eV[1]). Furthermore, both

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of Ba and Si are earth-abundant and suitable for large-scale production. Fundamental studies clarified various promising properties to certify its potential as the absorption layer of thin film solar cells. These include long diffusion length (10 μm [3]), long minority-carrier lifetime (14 μs [4]), wide range control of electron and hole densities [5-7], application of MoO_x hole transport layer to undoped n-type $BaSi_2$ [8], proposal of tin sulfide/$BaSi_2$ heterojunction solar cell to have a similar band diagram of pn homojunction [9], and so on.

The most of these fundamental studies have been done using $BaSi_2$ epitaxial films grown on Si(111) or Si(100) substrates by molecular beam epitaxy [10-12]. As for practical application, development of alternative method to permit realization of large-scale homogeneous $BaSi_2$ thin film is demanded. For this purpose, polycrystalline $BaSi_2$ was fabricated on glass substrate by radio-frequency sputtering [13, 14]. In our previous study, we proposed to utilize vacuum evaporation method, which enables us to fabricate films simply, quickly and stably. In fact, we have shown that single-phase $BaSi_2$ films can be formed on Si(111) substrate [15] and alkali-free glass substrate [16] by this method. In addition, the mechanism of film growth was discussed and we found that the vapor flux is Ba-rich and the supply of Si to the film is important to obtain homogeneous $BaSi_2$. Then, we speculated that high substrate temperature (≥500 ˚C) wound be necessary to fabricate $BaSi_2$ film. However, high substrate temperatures set a limit to choice of the substrate. It would be useful if we could somehow decrease the substrate temperature based on fundamental understanding of formation mechanisms.

In this paper, we report on the impact of the film thickness, substrate temperature and post-growth annealing times on the formation of $BaSi_2$ thin film on Si substrate grown by vacuum evaporation using $BaSi_2$ granules as source materials. These three factors were found to affect the composition in the film, and the oxidation was found to occur if the diffusion of Ba and Si is not enough. We revealed that appropriate choice of substrate temperature, film thickness, and post-growth *in-situ* annealing is necessary to realize homogeneous $BaSi_2$ thin film.

2. Experimental procedure

The films were deposited by vacuum evaporation under heating to form stoichiometric $BaSi_2$ film. 20×20-mm^2 Floating Zone n-type Si (111) (ρ>1000 Ω·cm) wafers were used for substrates after cleaning with 1% HF solution to remove SiO_2 on the surface. $BaSi_2$ (99% in purity, Kojundo Chemical Lab.) granules were used as sources and melted by heating for approximately 2 minute. The film thickness was varied by changing the source weight. The distance between source and substrate was set as 19 cm. The base pressure was lower than 1×10^{-5} mbar.

The grown films were characterized by Raman spectroscopy (Nanofinder, Tokyo Instruments, Inc.) with Ar^+ ion laser (λ= 488 nm), energy dispersive X-ray spectrometry (EDX), and scanning electron microscopy (SEM; JSM-7001FA, JEOL).

3. Results and discussion

Figure 1 shows cross-sectional SEM images of samples deposited under various conditions. Figures 1 (a) and (b) show the film deposited at 600 and 400 ˚C, respectively. Both of the films were fabricated by using the same weight of source. The film deposited at 600 ˚C is dense and the surface is flat. On the other hands, the film deposited at 400

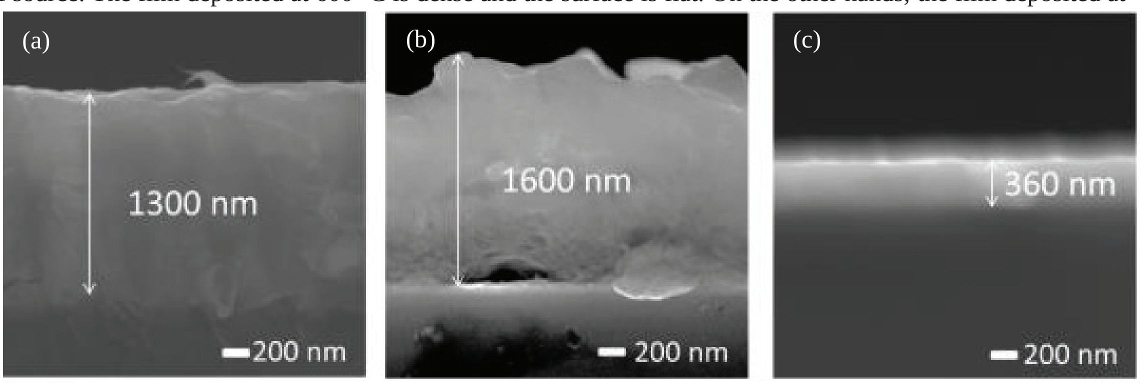

Fig. 1. Cross-sectional SEM images of the film grown at (a) 600 and (b), (c) 600 ˚C.